



**PATENT APPLICATION**

**IN THE UNITED STATES PATENT AND TRADEMARK OFFICE**

In re application of

Docket No: Q386121

Munehisa FUJITA, et al.

Appln. No.: 08/915,683

Group Art Unit: 1752

Confirmation No.: 1847

Examiner: WALKE, Amanda C.

Filed: August 21, 1997

For: **DIRECT POSITIVE PHOTOGRAPHIC SILVER HALIDE EMULSION AND  
COLOR PHOTOGRAPHIC LIGHT-SENSITIVE MATERIAL COMPRISING SAME**

**SUBMISSION OF EXECUTED DECLARATION UNDER 37 C.F.R. §1.132**

Commissioner for Patents  
Washington, D.C. 20231

Sir:

Submitted herewith is an executed Supplemental Declaration Under 37 C.F.R. §1.132  
signed by Takefumi HARA.

Respectfully submitted,

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Date: September 18, 2002

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For: DIRECT POSITIVE PHOTOGRAPHIC SILVER HALIDE EMULSION AND  
COLOR PHOTOGRAPHIC LIGHT-SENSITIVE MATERIAL COMPRISING  
SAME

SUPPLEMENTAL DECLARATION UNDER 37 C.F.R. §1.132

Assistant Commissioner for Patents  
Washington, D.C. 20231

Sir:

I, Takefumi HARA, do hereby declare and state as follows:

I am the same declarant who performed the experimentation described in the Declaration Under 37 C.F.R. §1.132 signed on April 22, 2002 and filed on April 25, 2002. My personal history remains as stated in my prior Declaration.

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In order to demonstrate superiority of the present invention over the prior art I have conducted the following experimentation.

#### EXPERIMENTATION

This experimentation was conducted in order to demonstrate the effect of the narrow distribution in thickness of the grains.

##### Preparation of Emulsion 1

The same procedure as for Emulsion A in Example of Evans (U.S. Patent 4,504,570) was performed to prepare a silver bromoiodide core emulsion.

The emulsion was a tabular grain emulsion. The tabular grains accounted for 90 % or more of all the silver halide grains as calculated in terms of projected area. The average of diameters of circles corresponding to projected areas of the tabular grains was 0.9  $\mu\text{m}$ , the average grain thickness was 0.1  $\mu\text{m}$ , and the average aspect ratio was 9:1.

The emulsion was subjected to chemical sensitization

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in the same manner as for the core grain of Emulsion No. D6 in Example 1 in the specification of the present application, except for changing the amount of 3,6-dithia-1,8-octanediol added to 0.04 g. That is, to the emulsion thus obtained were then added water, potassium bromide and deionized gelatin. The emulsion was then heated to a temperature of 75°C. To the emulsion were then added 0.04 g of 3,6-dithia-1,8-octanediol, 10 mg of Compound (1-16), 2.4 ml out of an aqueous solution obtained by dissolving 90 mg of potassium tetrachloroaurate and 1.2 g of potassium bromide in 1,000 ml of water, and 15 mg of lead acetate (in the form of aqueous solution). The emulsion was then heated to a temperature of 75°C for 180 minutes so that it was subjected to chemical sensitization.

An external shell was then formed on the grains in the same manner as for the core grain of Emulsion No. D6 in Example 1 in the specification of the present application, except that to the core grains thus chemically sensitized were then added

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the aqueous solution of silver nitrate and the aqueous solution of potassium bromide by the double jet process while the pBr value thereof was being kept to 2.8.

The emulsion was then washed by an ordinary flocculation method. The surface of the grains was subjected to chemical sensitization in the same manner as in Example 1 in the specification of the present application to obtain Emulsion 1.

The thus obtained emulsion was a tabular grain emulsion. The average of diameters of circles corresponding to projected areas of the tabular grains was  $2.5\text{ }\mu\text{m}$ , the average grain thickness was  $0.26\text{ }\mu\text{m}$ , and the average aspect ratio was 9.6:1. The tabular grain emulsion had the grain size distribution (coefficient of variation) of 25 %.

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Preparation of Emulsions 2 to 12

The same procedure as for Emulsion 1 was performed, except for changing an amount of 3,6-dithia-1,8-octanediol (hereinafter referred to as Compound A) at the formation of an external shell of the core grain, the kind of sulfur sensitizing agent at the chemical sensitization (Compound 1-16, 2-3 or 3-5) and pBr at the formation of the external shell as shown in Table 1 below to prepare Emulsions 2 to 12.

Preparation of Emulsion 13

In the Emulsion 1 above, the core grain was changed to silver bromide emulsion in the same manner as in the comparative experimentation in the Declaration under 37 C.F.R. §1.132 signed by Mr. MATSUNAGA on February 25, 1998 and filed on March 4, 1998. The thus obtained emulsion was subjected to chemical sensitization, external shell formation, flocculation, surface chemical sensitization in the same manner as for Emulsion 1 above, except for changing the amount of Compound A added and pBr at

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the formation of external shell as shown in table 1 below to prepare Emulsion 13.

The thus obtained emulsion was a tabular grain silver bromide emulsion. The average of diameters of circles corresponding to projected areas of the tabular grains was 2.5  $\mu\text{m}$ , the average grain thickness was 0.26  $\mu\text{m}$ , and the average aspect ratio was 9.6:1. The tabular grain emulsion had the grain size distribution (coefficient of variation) of 25 %.

Preparation of Emulsions 14 to 24

The same procedure as in the Emulsion 13 was performed, except for changing the kind of sulfur sensitizing agent at chemical sensitization of core grain, the amount of Compound A added and pBr at the formation of external shell as shown in table 1 below to prepare Emulsions 14 to 24.

The thickness a and b of shell of grain obtained in Emulsions 1 to 24 and distribution of thickness of the grains (the coefficient of variation) are shown in Table 1 below. The amount

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of Compound A added is an amount at the formation of external shell.

TABLE 1

Emulsion No.	Silver halide composition	Amount of Compound A added (g)	pBr	Compound No.	a (μm)	b (μm)	Distribution in thickness of the grains
1 (Comparison)	AgBrI	→ 0.00	2.8	1-16	0.71	0.08	15%
2 (Comparison)	AgBrI	0.17	2.5	1-16	0.72	0.08	28%
3 (Comparison)	AgBrI	→ 0.27	2.3	1-16	0.72	0.08	33%
4 (Comparison)	AgBrI	0.34	1.9	1-16	0.71	0.08	43%
5 (Comparison)	AgBrI	0.00	2.8	2-3	0.71	0.08	15%
6 (Comparison)	AgBrI	0.17	2.5	2-3	0.72	0.08	28%
7 (Comparison)	AgBrI	0.27	2.3	2-3	0.72	0.08	33%
8 (Comparison)	AgBrI	0.34	1.9	2-3	0.71	0.08	43%
9 (Comparison)	AgBrI	0.00	2.8	3-5	0.71	0.08	15%
10 (Comparison)	AgBrI	0.17	2.5	3-5	0.72	0.08	28%
11 (Comparison)	AgBrI	0.27	2.3	3-5	0.72	0.08	33%
12 (Comparison)	AgBrI	0.34	1.9	3-5	0.71	0.08	43%
13 (Invention)	AgBr	0.02	2.7	1-16	0.71	0.08	15%
14 (Invention)	AgBr	0.21	2.4	1-16	0.72	0.08	28%
15 (Comparison)	AgBr	0.29	2.2	1-16	0.72	0.08	33%
16 (Comparison)	AgBr	0.36	1.8	1-16	0.71	0.08	43%
17 (Invention)	AgBr	0.02	2.7	2-3	0.71	0.08	15%
18 (Invention)	AgBr	0.21	2.4	2-3	0.72	0.08	28%
19 (Comparison)	AgBr	0.29	2.2	2-3	0.72	0.08	33%
20 (Comparison)	AgBr	0.36	1.8	2-3	0.71	0.08	43%
21 (Invention)	AgBr	0.02	2.7	3-5	0.71	0.08	15%
22 (Invention)	AgBr	0.21	2.4	3-5	0.72	0.08	28%
23 (Comparison)	AgBr	0.29	2.2	3-5	0.72	0.08	33%
24 (Comparison)	AgBr	0.36	1.8	3-5	0.71	0.08	43%

The same procedure as for sample 101 in Example 1 in the specification of the present invention was performed except that the emulsions incorporated in the 8th layer, 15th layer



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and 22nd layer were replaced by these emulsions 1 to 24 respectively as shown in Table 2 below to prepare photosensitive elements (Sample 1 to 24). These samples were each processed in the same manner as in Example 1 in the specification of the present invention and evaluated for Dmax (maximum density), Dmin (minimum density), middle sensitivity and negative sensitivity in the same manner as in Example 1 in the specification of the present invention. The thus obtained results are shown in Table 3.

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TABLE 2

Sample No.	Emulsion incorporated in 8th layer	Emulsion incorporated in 15th layer	Emulsion incorporated in 22nd layer
1 (Comparison)	Emulsion 1	Emulsion 1	Emulsion 1
2 (Comparison)	Emulsion 2	Emulsion 2	Emulsion 2
3 (Comparison)	Emulsion 3	Emulsion 3	Emulsion 3
4 (Comparison)	Emulsion 4	Emulsion 4	Emulsion 4
5 (Comparison)	Emulsion 5	Emulsion 5	Emulsion 5
6 (Comparison)	Emulsion 6	Emulsion 6	Emulsion 6
7 (Comparison)	Emulsion 7	Emulsion 7	Emulsion 7
8 (Comparison)	Emulsion 8	Emulsion 8	Emulsion 8
9 (Comparison)	Emulsion 9	Emulsion 9	Emulsion 9
10 (Comparison)	Emulsion 10	Emulsion 10	Emulsion 10
11 (Comparison)	Emulsion 11	Emulsion 11	Emulsion 11
12 (Comparison)	Emulsion 12	Emulsion 12	Emulsion 12
13 (Invention)	Emulsion 13	Emulsion 13	Emulsion 13
14 (Invention)	Emulsion 14	Emulsion 14	Emulsion 14
15 (Comparison)	Emulsion 15	Emulsion 15	Emulsion 15
16 (Comparison)	Emulsion 16	Emulsion 16	Emulsion 16
17 (Invention)	Emulsion 17	Emulsion 17	Emulsion 17
18 (Invention)	Emulsion 18	Emulsion 18	Emulsion 18
19 (Comparison)	Emulsion 19	Emulsion 19	Emulsion 19
20 (Comparison)	Emulsion 20	Emulsion 20	Emulsion 20
21 (Invention)	Emulsion 21	Emulsion 21	Emulsion 21
22 (Invention)	Emulsion 22	Emulsion 22	Emulsion 22
23 (Comparison)	Emulsion 23	Emulsion 23	Emulsion 23
24 (Comparison)	Emulsion 24	Emulsion 24	Emulsion 24

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**TABLE 3**

Sample No.	Dmax			Dmin			Middle Sensitivity			Negative Sensitivity		
	Y	M	C	Y	M	C	Y	M	C	Y	M	C
1 (Comparison)	2.01	2.11	2.26	0.28	0.30	0.41	95	94	91	109	114	116
2 (Comparison)	1.97	2.08	2.22	0.30	0.32	0.43	93	91	89	112	117	119
3 (Comparison)	1.95	2.05	2.19	0.31	0.32	0.46	90	88	86	113	118	123
4 (Comparison)	1.87	1.98	2.06	0.35	0.38	0.49	87	83	82	115	121	126
5 (Comparison)	2.00	2.11	2.25	0.29	0.32	0.42	96	93	92	110	116	117
6 (Comparison)	1.98	2.07	2.20	0.31	0.34	0.44	94	90	88	113	118	120
7 (Comparison)	1.93	2.03	2.17	0.32	0.34	0.47	89	88	85	114	119	124
8 (Comparison)	1.87	1.97	2.06	0.37	0.39	0.51	86	84	79	116	122	127
9 (Comparison)	1.99	2.09	2.22	0.27	0.29	0.40	98	95	93	110	116	116
10 (Comparison)	1.96	2.05	2.19	0.30	0.31	0.44	95	93	90	113	118	120
11 (Comparison)	1.93	2.08	2.16	0.32	0.33	0.45	93	91	90	115	120	124
12 (Comparison)	1.92	2.04	2.13	0.36	0.37	0.48	88	85	83	117	122	128
13 (Invention)	2.18	2.32	2.48	0.15	0.16	0.23	111	128	149	82	82	81
14 (Invention)	2.14	2.29	2.45	0.17	0.18	0.26	109	125	147	85	88	87
15 (Comparison)	2.12	2.26	2.42	0.19	0.20	0.28	105	121	142	91	93	94
16 (Comparison)	2.06	2.21	2.35	0.23	0.24	0.32	102	119	138	99	101	103
17 (Invention)	2.16	2.30	2.47	0.14	0.14	0.21	110	126	147	82	81	81
18 (Invention)	2.13	2.27	2.43	0.17	0.17	0.24	108	124	145	86	86	87
19 (Comparison)	2.09	2.23	2.38	0.20	0.21	0.28	105	119	140	92	91	93
20 (Comparison)	2.04	2.19	2.33	0.25	0.26	0.33	101	117	137	100	102	101
21 (Invention)	2.18	2.31	2.48	0.15	0.15	0.22	113	129	148	84	82	82
22 (Invention)	2.14	2.30	2.44	0.18	0.18	0.25	109	125	146	88	89	88
23 (Comparison)	2.11	2.25	2.41	0.20	0.19	0.27	106	122	143	92	93	94
24 (Comparison)	2.05	2.19	2.36	0.24	0.25	0.33	103	118	139	101	101	103

As can be seen from the results shown in Table 3, samples 13 and 14, samples 17 and 18 and samples 21 and 22 according to the present invention are higher in Dmax, lower in Dmin, higher in middle sensitivity and unexpectedly remarkably lower in

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negative sensitivity than those of comparative samples 1 and 2, comparative samples 5 and 6, and comparative samples 9 and 10, respectively (i.e., a comparison between samples 13 and 14 according to the present invention and comparative samples 1 and 2, a comparison between samples 17 and 18 according to the present invention and comparative samples 5 and 6, and a comparison between samples 21 and 22 according to the present invention and comparative samples 9 and 10). Furthermore, when samples having different distributions (coefficient of variation) in thickness of the grains (i.e., samples having the distribution in thickness of the grains of not more than 30% and samples having the distribution in thickness of the grains of more than 30%) were compared, in case where the distribution is not more than 30%, the AgBr emulsion samples according to the present invention are unexpectedly remarkably large in particular in an effect inhibiting negative sensitivity of Y, M and C as compared with AgBrI emulsion samples for comparison

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(i.e., a comparison of sample 14 according to the present invention and comparative sample 15 vs. comparative samples 2 and 3, a comparison of sample 18 according to the present invention and comparative sample 19 vs. comparative samples 6 and 7, and a comparison of sample 22 according to the present invention and comparative sample 23 vs. comparative samples 10 and 11).

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I declare further that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that the statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issuing therefrom.

Date: September 2, 2002

Takefumi Hara  
Takefumi HARA